NEUTRON IRRADIATION EFFECTS ON SPARK PLASMA SINTERED BORON CARBIDE

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In this study, spark plasma sintered boron carbide (B_4C) was examined against neutrons. The specimens were irradiated by reactor neutrons (include both thermal and fast neutrons) up to fluence of $1.37x10^{21} n{\cdot}m^{-2}$. Thermal and fast neutrons cause swelling by different interactions with boron (^{10}B) atoms in the related materials. X-Ray diffraction (XRD) patterns and scanning electron microscopy (SEM) images were investigated for initial and irradiated samples. In addition, lattice parameters and unit cell volumes were calculated for the samples. The swelling percentages were calculated to be within a range of 0.49-3.80 % (average 1.70 %) for the outer surface of the materials for applied neutron irradiation doses.

Introduction

Boron carbide (B_4C) is extremely important material for nuclear technology because of its advanced properties such as high hardness, high melting point, low density and high thermal neutron capture cross-section [1, 2]. Boron carbide is widely used in fission reactors as control rods and have potential to be used in fusion reactors [2-4]. Since boron carbide and regarding materials are commonly used in nuclear applications, radiation effects on boron carbide is one of the most important subjects that has to be clearly understood. Therefore, there are remarkable studies on irradiation effects on boron carbide and related materials in the literature [3-5].

Spark plasma sintering (SPS) technique is a relatively new process that enables high heating rate, low sintering temperature which prevents grain coarsening during sintering since all of the sintering process takes place within minutes rather than hours or days compared to other sintering methods.

Either thermal neutrons (E<0.025 eV) or fast neutrons (E>0.1 MeV) affect the structure of the materials; in the central thimble or the reactor. Thermal neutrons mostly have inelastic interaction (n,α) with the ^{10}B atoms which have reasonably high neutron absorption cross-section (3855 barns) [25], in B_4C-Al composites. The remaining helium causes the swelling for boron related materials [6-8]. On the other hand elastic collisions are leading to fast neutrons and material-atoms interactions. Fast neutrons can cause displacement of lattice atoms from their original places which cause swelling with change in the lattice parameters and cell volumes of the materials [9].

In this work, the reactor neutrons irradiation effects on the spark plasma sintered monolithic boron carbide (B_4C) were investigated. The effects of reactor neutrons on the structure of boron carbide were evaluated.

X-Ray Diffraction (XRD) and SEM (Scanning Electron Microscopy) were carried out for initial and irradiated boron carbide-aluminum composites. The phase composition of the samples was carried out by XRD where the surface morphology was analyzed by means of SEM. The structure features of boron carbide were carried out. The lattice parameters of crystalline and unit cell volumes were investigated for B_4C for initial and reactor neutrons irradiated samples.

Experimental

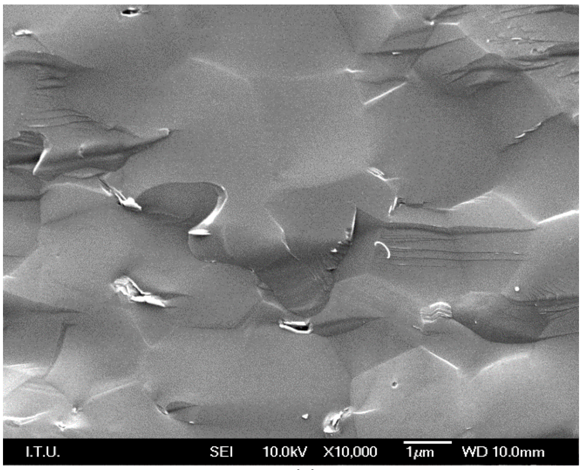
Boron carbide-aluminum composites were produced by spark plasma sintering technique. Commercial HS grade B_4C powders from H.C. Starck Co., with an average particle size of 0.7 µm (99.5% purity) were used to produce B_4C composites. Samples having 50x50 mm cross section and 5mm height for B_4C composites were sintered in SPS and their behaviors under reactor neutrons were analyzed for these dimensions for the first time in literature.

The samples were sintered by using the SPS apparatus (SPS-7.40MK-VII, SPS Syntex Inc.). After initial pressure of 10 MPa was applied manually for compaction before sintering, 100°C/min heating rate was used with 40 MPa of applied pressure from room temperature to sintering. An optical pyrometer was used for measuring the temperature from the outer side of the die. All of the samples were subjected to 4 minute soaking time. Whole process was carried out in vacuum and shrinkage, displacement, temperature, vacuum, current, and voltage for every 5 seconds were recorded. At the end of the process, sintered B_4C was obtained. Before irradiation, sintered samples were sand blasted and cut into required pieces so as to determine their density values by Archimedes principle.

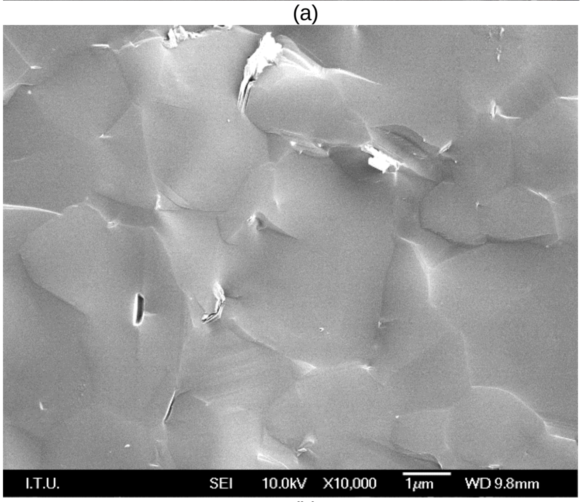
Samples were irradiated in the central thimble of Istanbul Technical University TRIGA Mark-II Research Reactor with a power of 160 kW. A spectrum of neutron fluxes at different energies (up to 10 MeV) which include thermal (E<0.025 eV) and fast (E>0.1 MeV) neutrons in the central thimble of the reactor [10]. The average total neutron flux was $6.34{\cdot}10^{16} n{\cdot}m^{-2}{\cdot}s^{-1}$. The samples were irradiated to total neutron (both thermal and fast neutrons) fluences up to $1.37x10^{21} n{\cdot}m^{-2}$. However, the main subject which we focused on was the swelling of the materials after neutron irradiation. The swelling effect of neutrons comes from both thermal and fast neutrons which have some different interaction mechanisms on boron atoms [6, 11].

Results and Discussion

SEM analysis of initial and irradiated samples were figured out and compared with each other. SEM images of the initial and neutron irradiated samples were given in Fig. 1.



(a)



(b)

Fig. 1. SEM images of initial (a) and irradiated by $1.37x10^{21}n.m^{-2}$ neutron fluence (b) of B_4C.

From the Fig. 1, it could be said that no significant change on the boron carbide due to applied neutron fluences. In addition XRD-patterns were analyzed for spark plasma sintered boron carbide as initial and after irradiated by reactor neutrons. The phase composition of the irradiated samples were compared with initial ones and given in Fig. 2.

There was no new phase composition which could be related with the applied neutron irradiations. However, little peak shifts were observed which were caused by irradiation for boron carbide. Therefore *d* spacing values between the planes were increased with neutron irradiation of the samples. The lattice parameters and unit cell volumes were carried out by using hexagonal structure as a reference [7, 12] and obtained results were given in Table 1.

The lattice parameter-neutron fluence graphs were drawn for B_4C phase of the materials by using the values in Table 1 and graphs were shown in Fig. 3.

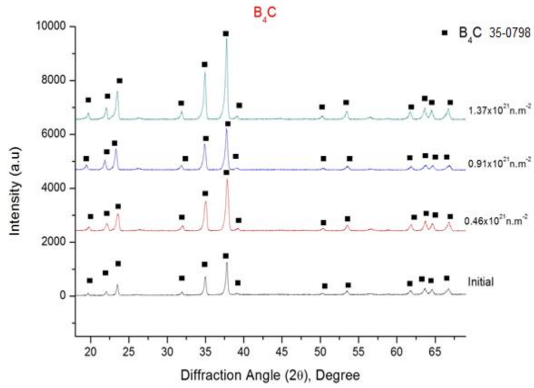


Fig. 2. XRD patterns of initial and neutron irradiated boron carbide.

Table 1. The lattice parameters and unit cell volumes of boron carbide for initial and irradiated samples.

Neutron Fluence ($10^{21}n{\cdot}m^{-2}$)	The Lattice Parameters (A°)		Cell Volume (A°^{3})	Swelling (%)
	a	*c*		
0	5.6045	12.0002	326.4375	-
0.46	5.6001	12.0786	328.0473	0.4931
0.91	5.5957	12.4959	338.8431	3.8003
1.37	5.6090	12.0789	329.1017	0.8161

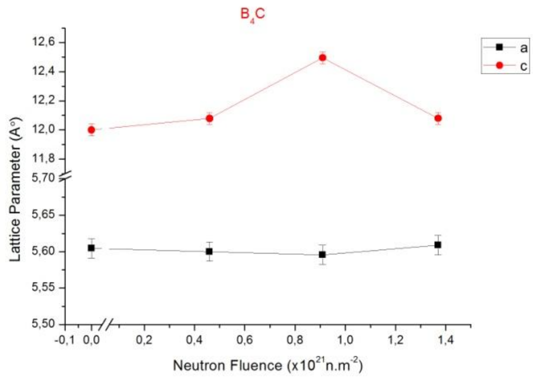


Fig. 3. The lattice parameters of the initial and neutron irradiated boron carbide.

From Fig. 3, it could be seen the lattice parameter *a*, doesn't change meaningfully with the neutron irradiation for boron carbide. On the other hand the lattice parameter *c* slightly increases with increasing neutron fluence. The propagated relative errors were calculated in a range of 0.4-0.7 percentage. Furthermore, the lattice parameters were calculated from surface of the irradiated samples. Cell volume graphs of the samples were drawn against irradiation fluences and given in Figure 4.

The swelling percentages of spark plasma sintered boron carbide were obtained to be in the range of 0.49-3.80. Although there were differences in production and irradiation conditions, it could be said that our results were compatible with the literature

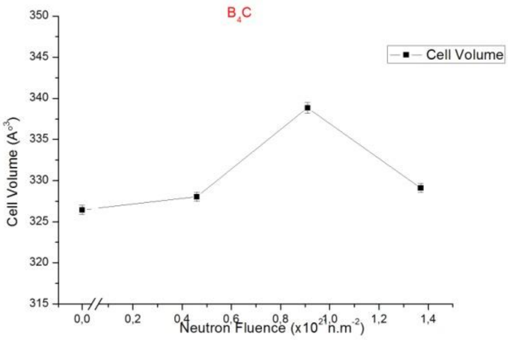


Fig. 4. The cell volumes of B_4C for boron carbide.

[9]. In addition, the swelling was one of the important parameters for B_4C which is essential for nuclear applications. There were some researches and new equipment for the reactor materials such as swelling resistant control rods for the nuclear reactors and applications [13].

Conclusions

Spark plasma sintered boron carbide for the first time were produced having 50x50x5 mm^3 dimensions were investigated under reactor neutrons up to $1.37 \times 10^{21} n \cdot m^{-2}$ fluence. No new phase formation, or phase transition could be observed which could be related with the neutron irradiation. In addition, there wasn't significant change on lattice parameter *a* due to neutron irradiation. However, the lattice parameter *c* and cell volumes of the samples were increased with neutron irradiation. Swelling was observed in boron carbide structure which is caused by both thermal and fast neutrons. The average swelling percentage was obtained as 1.70 %. The differences maybe caused from the irradiation position in the central thimble of the reactor.

Further researches need to make theoretical calculation and partition of the thermal and fast neutron irradiation effects on the whole material by including burn up (depletion of ^{10}B in time) parameter. In conclusion, an experimental study was realized on spark plasma sintered boron carbide-aluminum composites which could be used in fission reactors (as control rods) and waste disposal applications and also for possible uses in fusion reactors.

Acknowledgements

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References

1. *Thevenot F., Key Eng*. // Mater. 1991. V. 56-57. P. 59.
2. *Kitamura J., Usuba S., Kakudate Y., et al* // Diam.and Relat. Mater. 2003. V. 12. P. 1891.
3. *Pilladi T. R., Panneerselvam G., Antonysamy S., Ganesan V.* // Ceram. Int. 2012. V. 38. P.3723.
4. *Simeone D., Mallet C., Dubuisson P., Baldinozzi G., Gervais C., Maquet J.* // J. of Nucl. Mater. 2000. V. 277. P.1.
5. *Maruyama T., Onose S., Kaito T., Horiuchi H. //* J. of Nucl. Sci.and Technol. 1997. V. 34. P. 1006.
6. *Monterrosa A., Iyengar A., Huynh A., Madaan C*. // Rep. UCBTH-12-007. 2012.
7. Kharisov *B.I., Kharissova O. V., Mendez U.O.* Radiation Synthesis of Materials and Composites, CRC Press, Florida, 2013.
8. *Mohnaty R.M., K. Balasubramanian, S.K. Seshadri R.M.* Evolution of Crystallographic Structures andPhases in Micropyretically FormedBoron Rich Boron Carbide –a New Material System in: C.Sikalidis(Ed) Advances in Ceramics, Intech, 2011.
9. *Zinkle S.J., Kinoshita C. //* J. of Nucl. Mater. 1997. V. 251. P. 200.
10. *Gebhardt O., Gavillet D.* // J. of Nucl. Mater. 2000. V. 279. P. 368.
11. *Kwon, J. and Motta, A. T*., "Role of Radiation in BWR Core Shroud Cracking, in: Reactor Dosimetry, ASTM STP 1398, in: J. G. Williams, D. W. Vehar, F. H. Ruddy and D. M. Gilliam, (Eds.), American Society for Testing and Materials, West Conshohocken, PA. 2001. P. 607-616.
12. *Tucker C.W., Jr., Senio P*. // Acta Crystallogr. 1955. V. 8. P. 371.
13. IAEA-TecDoc-813, Advances in control assembly materials for water reactors, ISSN 1011-4289, IAEA, Vienna, 1995.